

# Technical Language Service

Translations From And Into Any Language

## **JAPANESE / ENGLISH TRANSLATION OF**

Japanese Patent Application JP 2003 - 191266 A

Resin Composition and Sheet/Film Obtained Using This Resin Composition

Your Ref: 103003 - 04

For: Eastman Chemical Company

9109 Eagle Hills Dr . Las Vegas NV 89134 . ph: 866-722-7990 (toll-free) . fax: 702-973-6009 . email: sales@tls-translations.com

(19) Japanese Patent Office (JP)

(11) Unexamined Patent Application No:

# (12) Unexamined Patent Gazette (A) Kokai 2001-191266 (D2001-101266A)

(P2001-191266A)

(43) Date of Publication: July 8, 2003

		·				(43) Date of 1 dollcation. July 8, 2003	
(51) Int. Cl. <sup>7</sup>		Class. Symbols	FI			Subject Codes (Reference)	
B 2	9 C 43/24		B 29 C	43/24		4F071	
C 0	8 J 5/18	CFD	C 08 J	5/18	CFD	4F204	
C 0	8 K 5/09		C 08 K	5/09		4J002	
C 0	8 L 67/02	•	C 08 L	67/02			
' // B 2	9 K 67:00		B 29 K	67:00		•	
Reque	st for Examination: N	lot yet submitted	Number of	Claims:	3	Total of 12 pages [in original]	
						(Continued on last page)	
(21)	Application No.:	2001-397378		(71)	Applicant:	000010010	
		(P2001-397378)			Lonseal Co	orporation	
(22)	Date of Filing:	December 27, 2001			15-3 Midori 4-chome, Sumida-ku, Tokyo-to		
				(72)	Inventor:	Toshio Ohama	
					c/o Lonsea	l Corp., Tsuchiura Office	
					5-3 Higash Ibaraki-ken	i Nakanuki-machi, Tsuchiura-shi,	
				(72)	Inventor:	Takeshi Watanabe	
					c/o Lonseal	Corp., Tsuchiura Office	
					5-3 Higashi Ibaraki-ken	Nakanuki-machi, Tsuchiura-shi,	
				(74)		Sadayuki Hosoi, Patent Attorney (and 2 others)	
						(Continued on last page)	

#### (54) [Title of the Invention]

# Resin Composition and Sheet/Film Obtained Using This Resin Composition

#### (57) [Summary]

[Object] To provide a polyester resin composition for calendering in which the transparency that is a characteristic of non-crystalline polyester resins can be maintained, which is easy to release from a metal roll and is virtually devoid of plating out onto the metal rolls, and in which longitudinal streaks are prevented from being generated as a result of the entrainment of air in the sheet or film obtained by means of calendering.

[Means of Achievement] 0.2 to 3.0 weight parts of a fatty acid with a carbon number of 20 or higher and 0.01 to 3.0 weight parts of a metal salt of a fatty acid are admixed in 100 weight parts of a polyester resin consisting of repeating units of a dicarboxylic acid component and a glycol component.

#### [Claims]

[Claim 1] A polyester resin composition used for calendering, characterized in that 0.2 to 3.0 weight parts of a fatty acid with a carbon number of 20 or higher and 0.01 to 3.0 weight parts of a metal salt of a fatty acid are admixed into 100 weight parts of a polyester resin comprising repeating units of a dicarboxylic acid component consisting of 80 mol% or more of a mixture of one, two, or more types selected from terephthalic acid, naphthalene dicarboxylic acid, 1,4-cyclohexane dicarboxylic acid, and isophthalic acid, as well as a glycol component consisting of 80 mol% or more of a mixture of one, two, or more types selected from ethylene glycol, neopentyl glycol, diethylene glycol, and 1,4-cyclohexane dimethanol.

[Claim 2] The polyester resin composition used for calendering according to claim 1, wherein the admixed quantity of the metal salt of a fatty acid is the same or less than the admixed quantity of the fatty acid.

[Claim 3] A sheet/film molded by means of calendering using the polyester resin composition according to claims 1 or 2.

### [Detailed Description of the Invention]

#### [0001]

[Technological Field of the Invention] The present invention relates to a polyester resin composition used for calendering, and to a sheet or film obtained by means of calendering the resin composition.

#### [0002]

[Prior Art] Polyester resin compositions, particularly crystalline polyethylene terephthalate resin (hereinafter referred to as PET) made up of repeating units of terephthalic acid and ethylene glycol, have been developed as materials used for molding in manufactured products having various applications, and among these applications, products molded into a sheet or film are widely used as packaging material for food and pharmaceutical items, covers used for containers,

raw material used in blister packages, and as lamination sheeting for construction, consumer electronics, and automobiles.

[0003] Sheets that are molded using PET resin are generally manufactured by means of an extrusion molding method. The extrusion molding method consists of a molding method whereby a molten resin is discharged after adjusting the dies so as to produce a sheet with a specific thickness, and the molten resin is taken off while being rapidly cooled to a temperature lower than the softening temperature thereof. Therefore, release from the takeoff rolls is easy, but because portions of differing thickness can form in parts of the sheet even if the dies are adjusted, which reduces the accuracy of the sheet thickness, this can cause trouble in printing, lamination, coating, and other fabrication processes, and holes sometimes form in the sheet during secondary molding such as vacuum/pressure forming. In addition, the molding speed is low during molding of the sheet by means of extrusion molding, and the productivity thereof can hardly be considered good.

[0004] For such reasons as these, calendering methods are often applied in the manufacture of sheets and films, as opposed to extrusion molding methods. Calendering is a molding method whereby a molten resin is rolled with heated metal rolls (calender rolls) in order to manufacture a sheet or film with a prescribed thickness, and this method can yield a product having good accuracy in terms of the thickness of the molded sheet/film and excellent quality without the drawbacks that usually occur near the dies in extrusion molding, and because of its high molding speed and productivity, this method is suitable for manufacturing large quantities of products that have the same specifications.

[0005] However, it has been nearly impossible to apply crystalline PET resin to calendering in order to manufacture sheets and films. This is because crystalline PET resin has a high melting point, and depending on the limit for the temperature setting of the calendering machine, the melt tension may become extremely low when a melt is formed at a temperature at or above the melting point thereof, making such a resin inapplicable to calendering, which requires high melt tension.

[0006] Consequently, research and development have been conducted regarding PET resins with the aim of changing the chemical structure of the resins in order to alter their workability and physical properties. Specifically, the crystallinity of a PET resin can be controlled and part of the resin can be rendered completely non-crystalline by substituting a portion of the terephthalic acid

and ethylene glycol monomer components thereof with another component, and performing random copolymerization. Controlling crystallinity in this manner enables improvement of physical properties such as high transparency, and has made it possible to apply such a resin to calendering.

[0007] However, because of the significant adhesion of highly polar resins such as PET resin to the heated metal rolls (calender rolls), using such resins as resin elements in calendering becomes impossible, and the addition of various additives has been proposed for facilitating release from the metal rolls. Examples of such proposals are disclosed in JP (Kokai) No. 11-343353, 2000-186191, 2000-302951, 2000-327891, 2001-64496, and the like. Products that have low affinity to the resin are generally considered to be effective as additives for facilitating release from the metal rolls. However, because the transparency of the molded sheet/film declines, among other such effects, which causes the resin to plate out on the metal rolls when such additives are admixed therein, modifying the resin by controlling its crystallinity to enable application thereof to calendering is only slightly successful in allowing the resin to be readily released from the metal rolls while at the same time preventing plating out to the metal rolls without compromising transparency in PET-type polyester resins.

#### [0008]

[Problems to Be Solved by the Invention] An object of the present invention, which was developed in view of the foregoing drawbacks, is to provide a polyester resin composition used for calendering that is still allowed to maintain the transparency that is a characteristic of PET-type polyester resins, particularly a polyester resin whose crystallinity is controlled (non-crystalline polyester resin), which is easy to release from a metal roll (calender roll) with minimal plating out to the metal rolls, and in which longitudinal streaks are prevented from being generated due to entrainment of air in the sheet or film obtained by means of calendering; and to provide a sheet or film obtained by means of calendering the polyester resin composition.

#### [0009]

[Means Used to Solve the Above-Mentioned Problems] As a result of concentrated investigation aimed at overcoming the abovementioned drawbacks, the inventors discovered that the abovementioned drawbacks can be overcome by means of a polyester resin composition used

for calendering in which specific additives are admixed in specific quantities into a specific polyester resin to form a resin composition, and thus developed the present invention.

[0010] Specifically, the polyester resin composition pertaining to the present invention is characterized in that 0.2 to 3.0 weight parts of a fatty acid with a carbon number of 20 or higher and 0.01 to 3.0 weight parts of a metal salt of a fatty acid are admixed into 100 weight parts of a polyester resin comprising repeating units of a dicarboxylic acid component consisting of 80 mol% or more of a mixture of one, two, or more types selected from terephthalic acid, naphthalene dicarboxylic acid, 1,4-cyclohexane dicarboxylic acid, and isophthalic acid, as well as a glycol component consisting of 80 mol% or more of a mixture of one, two, or more types selected from ethylene glycol, neopentyl glycol, diethylene glycol, and 1,4-cyclohexane dimethanol. It is more preferable that the admixed quantity of the aforementioned metal salt of a fatty acid be the same or less than the admixed quantity of the aforementioned fatty acid. Also, the sheet/film pertaining to the present invention is molded by means of calendering using the previously described polyester resin composition.

#### [0011]

[Embodiments of the Invention] Preferred embodiments of the present invention will be described in detail hereinafter. The polyester resin used in the present invention is made up of repeating units of a dicarboxylic acid component consisting of 80 mol% or more of a mixture of one, two, or more types selected from terephthalic acid, naphthalene dicarboxylic acid, 1,4-cyclohexane dicarboxylic acid, and isophthalic acid, as well as a glycol component consisting of 80 mol% or more of a mixture of one, two, or more types selected from ethylene glycol, neopentyl glycol, diethylene glycol, and 1,4-cyclohexane dimethanol.

[0012] Also, a polyester resin is preferred for use that is easily obtained as a raw material, and that comprises repeating units of a dicarboxylic acid component containing at least 90 mol% or more of terephthalic acid, and a glycol component containing 10 to 95 mol% of 1,4-cyclohexane dimethanol and 90 to 5 mol% of ethylene glycol, and a non-crystalline polyester resin is suitable for use among such resins. The reason for this is that a non-crystalline polyester resin that can be melted at a low temperature is favorable for calendering, which requires a high melt tension, and a sheet or film that has good surface smoothness and transparency is obtained by means of calendering such a resin. The non-crystallinity of the polyester resin differs according to the composition ratio of its monomers, but in order to achieve non-crystallinity while obtaining good

moldability as a result of calendering, the resin preferably comprises 100 mol% of terephthalic acid, 30 to 90 mol% of ethylene glycol, and 10 to 70 mol% of 1,4-cyclohexane dimethanol, and more preferably comprises 100 mol% of terephthalic acid, 65 to 80 mol% of ethylene glycol, and 20 to 35 mol% of 1,4-cyclohexane dimethanol.

[0013] The polyester resin used in the present invention is obtained by means of reacting a dicarboxylic acid component that comprises terephthalic acid with a glycol component that comprises 1,4-cyclohexane dimethanol and ethylene glycol, and polycondensing the product in the presence of a specific catalyst. Specifically, a colorless transparent copolyester is obtained by means of a process whereby a dicarboxylic acid component comprising at least 90 mol% of terephthalic acid is reacted at a temperature sufficient for esterification or transesterification with a glycol component comprising 10 to 95 mol% of 1,4-cyclohexane dimethanol and 90 to 5 mol% of an ethylene glycol component, and the reacted product thus obtained is polycondensed for less than two hours at an absolute pressure lower than 1.333 kPa in the presence of a catalyst or inhibitor system made up of 0 to 75 ppm of Mn, 50 to 150 ppm of Zn, 5 to 20 ppm of Ti, 5 to 200 ppm of Ge, and 10 to 80 ppm of P, based on the weight of the copolyester.

[0014] Also, in the dicarboxylic acid component of the polyester resin used in the present invention, a mixture of one, two, or more types selected from terephthalic acid, naphthalene dicarboxylic acid, 1,4-cyclohexane dicarboxylic acid, and isophthalic acid accounts for at least 80 mol% or more thereof. The remaining dicarboxylic acid component of less than 20 mol% includes succinic acid, glutamic acid, adipic acid, sebacic acid, suberic acid, azelaic acid, dodecanedioic acid, fumaric acid, maleic acid, itaconic acid, orthophthalic acid, 2,6-naphthalene dicarboxylic acid, and the like. Furthermore, a small quantity of a tri- or tetracarboxylic acid may be admixed into the dicarboxylic acid component of less than 20 mol% to introduce long-chain branches into the molecular structure of the polyester resin. The melt tension can be increased as a result of the introduction of long-chain branches into the molecular structure of the polyester resin, which is advantageous for calendering. Also, trimellitic acid and the like may be included as the tricarboxylic acid, and pyromellitic acid and the like may be included as the tetracarboxylic acid.

<sup>&</sup>lt;sup>1</sup> Translator's note: The prefix "di" appears twice in succession in the original, which appears to be a typographical error.

[0015] On the other hand, a mixture of one, two, or more types selected from ethylene glycol, neopentyl glycol, diethylene glycol, and 1,4-cyclohexane dimethanol accounts for at least 80 mol% or more of the glycol component of the polyester resin used in the present invention. The remaining glycol component of less than 20 mol% includes, for example, propylene glycol, 1,3-propanediol, 2,4-dimethyl-2-ethylhexane-1,3-diol, 2,2-dimethyl-1,3-propanediol, 2-ethyl-2-butyl-1,3-propanediol, 2-ethyl-2-isobutyl-1,3-propanediol, 1,4-butanediol, neopentyl glycol, 1,5-pentadiol, 1,6-hexanediol, 1,8-octanediol, 2,2,4-trimethyl-1,6-hexanediol, thiodiethanol, 1,2-cyclohexane dimethanol, 1,3-cyclohexane dimethanol, 2,2,4,4-tetramethyl-1,3-cyclobutanediol, and the like. A small quantity of a tri- or tetrahydric alcohol may also be admixed into the glycol component of less than 20 mol% to introduce long-chain branches into the molecular structure of the polyester resin. In this case, trimethylol propane may be cited as an example of a trihydric alcohol, and pentaerythritol may be cited as an example of a tetrahydric alcohol.

[0016] There is no particular limit regarding the molecular weight of the polyester resin suitable for use in the present invention, but if the number-average molecular weight is less than 10,000, the melt tension of the molten resin is low, and the temperature range that enables molding in calendering is narrowed, whereas if the number-average molecular weight exceeds 200,000, the viscosity of the molten resin increases, which places an increased load on the calendering equipment and has an adverse effect on the smoothness of the sheet or film thus obtained. Consequently, a number-average molecular weight in a range of 10,000 to 200,000 is preferred for the purpose of reducing the load on the calendering equipment and maintaining a wide range of molding conditions, taking into account that a sheet or film having good surface smoothness needs to be obtained.

[0017] The molecular weight distribution of the polyester resin used in the present invention is also not particularly limited. However, when the high melt tension required in calendering and the low viscosity required for the purpose of reducing the load on the calendering equipment are sought at the same time, it is effective to widen the overall molecular weight, add small amounts of resins with extremely high molecular weights, or adopt other measures, as is generally practiced. These measures can sometimes be implemented by way of polymerization, but may also be implemented by adopting an approach in which polyester resins of the same type having different molecular weights are mixed together.

[0018] Also, polyester resins that can be used in the present invention include products that are already commercially available. For example, non-crystalline polyester resins made up of 100 mol% of terephthalic acid, 60 to 75 mol% of ethylene glycol, and 40 to 25 mol% of 1,4-cyclohexane dimethanol are marketed as Kodar PETG Copolyester grade Eastar PETG 6763 as well as Tsunami Copolyester grade GS1, GS2, GS3, and GS4 (Eastman Chemical Products, Inc.), and the like, and these products are suitable for use with the present invention. [0019] On the other hand, the fatty acid admixed into the polyester resin in the present invention is limited to fatty acids with a carbon number of 20 or higher. Specifically, because the fatty acid evaporates at a lower temperature with a reduction in the carbon number, if a fatty acid with a carbon number lower than 20 is admixed into the polyester resin, a large amount of evaporation occurs in the temperature range of 170°C to 220°C (which is the range that is suitable for calendering), making it more difficult to continuously release the resin from the metal rolls for long periods of time, causing the manner in which the resin is released from the metal rolls to vary with the conditions, and bringing about other drawbacks. It is therefore necessary that the fatty acid have a carbon number of 20 or above in order to make it easier for the resin to be released from the metal rolls in a stable manner for a long period of time within the temperature range suitable for calendering the polyester resin.

[0020] Fatty acids with a carbon number of 20 or above include, for example, behenic acid, tetracosanic acid, hexacosanic acid, heptacosanic acid, montanic acid, triacontanoic acid, dotriacontanoic acid, and the like. These fatty acids can be obtained as high-purity reagents, but are in many cases expensive, and are not generally used as additives in resin compositions. However, there are some fatty acids with a carbon number of 20 or above that are available at low cost as additives used for resins, such as montanic acid, and a fatty acid used as a resin additive comprising montanic acid is suitable for use in the present invention as well.

[0021] On the other hand, admixing a fatty acid with a carbon number of 20 or above in the polyester resin has the characteristic of reducing the glass transition temperature of the polyester resin. This indicates that a fatty acid with a carbon number of 20 or above is easily mixed with the polyester resin to be released from the metal is one that has low affinity to the resin and is difficult to mix therewith, and a fatty acid with a carbon number of 20 or above is easily mixed with the polyester resin, and is characterized in being capable of making it easier for the resin to

JP 2003 - 191266 A

be released from the metal. For this reason, characteristics are obtained whereby the molded sheet/film has excellent transparency and does not plate out to the metal rolls, regardless of how easy it is to release the resin from the metal rolls. Fatty acids with a carbon number of 20 or above differ from other release agents and lubricants for polyester resins in this respect.

[0022] When a fatty acid with a carbon number of 20 or above is admixed in the polyester resin, the admixed quantity thereof is in a range of 0.2 to 3.0 weight parts per 100 weight parts of the polyester resin. This is because if the admixed quantity of the fatty acid with a carbon number of 20 or above is smaller than 0.2 weight parts, it becomes difficult to release the resin from the metal rolls, and alternatively, if the admixed quantity exceeds 3.0 weight parts, the transparency of the sheet or film declines. Therefore, the admixed quantity of the fatty acid with a carbon number of 20 or above is limited in the present invention to a range of 0.2 to 3.0 weight parts per 100 weight parts of the polyester resin in order to be able to release the resin from the metal rolls with ease and in a stable manner during calender molding, while achieving high transparency in the sheet or film.

[0023] Furthermore, it is essential that a metal salt of a fatty acid be admixed in addition to the fatty acid with a carbon number of 20 or above in the polyester resin composition of the present invention. A sheet or film that is easily released from a metal roll and that has good transparency can be manufactured using a fatty acid with a carbon number of 20 or above if the admixed quantity thereof is specified, but because air is entrained in the mass of molten resin (usually referred to as "bank") formed between the metal rolls during calendering, and longitudinal streaks can appear on the surface of the sheet or film, it is effective to admix a metal salt of a fatty acid therewith to overcome such drawbacks.

[0024] In this case, the carbon number of the fatty acid component in the metal salt of the fatty acid is not particularly limited. Metal salts of fatty acids include, for example, lithium stearate, magnesium stearate, calcium stearate, potassium stearate, aluminum stearate, strontium stearate, barium stearate, cadmium stearate, zinc stearate, lead stearate, calcium laurylate, barium laurylate, cadmium laurylate, zinc laurylate, lithium myristate, zinc oleate, calcium behenate, zinc behenate, magnesium behenate, lithium behenate, calcium montanate, magnesium montanate, sodium montanate, cadmium 2-ethylhexoate, zinc 2-ethylhexoate, and the like. These metal salts of fatty acids include products that are commercially available for use as resin additives, which are suitable for use in terms of economy.

JP 2003 - 191266 A

[0025] The quantity in which the metal salt of a fatty acid is admixed in the polyester resin is preferably in a range of 0.01 to 3.0 weight parts per 100 weight parts of the polyester resin. Even a small quantity of the metal salt of a fatty acid is capable of overcoming the adverse effects brought about if a fatty acid alone is admixed as described above, but if the admixed quantity thereof is less than 0.01 weight parts, the desired effects are not obtained, and if the admixed quantity is over 3.0 weight parts, the transparency of the sheet/film thus molded is reduced. Therefore, an admixed quantity of the metal salt of a fatty acid that is in a range of 0.01 weight parts to 3.0 weight parts is preferred in order to maintain high transparency in the polyester resin while achieving the effects provided as a result of the joint use of the metal salt of a fatty acid. [0026] On the other hand, if the admixed quantity of the metal salt of a fatty acid is more than that of the fatty acid, the resin becomes somewhat more difficult to release from the metal rolls and it becomes more difficult to perform calendering under certain conditions, so it is preferable that the admixed quantity of the metal salt of a fatty acid be the same or less than that of the fatty acid in order to enable consistent calendering under a wide range of molding conditions, and to manufacture a sheet or film having a good outward appearance.

[0027] It is also sometimes effective to also admix a fatty acid ester or oxidized polyethylene wax into a polyester resin into which a fatty acid and a metal salt of a fatty acid have been jointly admixed in order to make it easier for the resin to be released from the metal rolls. However, because fatty acid esters and oxidized polyethylene waxes have significant effects in enhancing release properties, their use can lead to plating out and decreased resin transparency. Therefore, any admixed quantity thereof must be kept low. Specifically, it is preferable that the fatty acid or metal salt of a fatty acid be admixed in the same quantity or less, and that the admixed quantity thereof be 0.5 weight part or less.

[0028] The carbon number of the fatty acid component of the fatty acid ester used herein is not particularly limited. However, taking into account the volatility of the ester thus obtained, natural wax or a synthetic wax made up of an ester of a saturated fatty carboxylic acid with a carbon number of 12 to 28 and a saturated fatty alcohol with a carbon number of 2 to 30 is preferred. The saturated fatty carboxylic acid with a carbon number of 12 to 28 constituting the synthetic wax may include, for example, lauric acid, mysteric<sup>2</sup> acid, stearic acid, behenic acid, lignoceric

<sup>&</sup>lt;sup>2</sup> Translator's note: May be a typographical error for "myristic." This term is corrected elsewhere in the translation.

acid, cerotic acid, montanic acid, and the like, and the saturated fatty alcohol with a carbon number of 2 to 30 may include, for example, ethyl alcohol, octyl alcohol, lauryl alcohol, stearyl alcohol, behenyl alcohol, pentacosyl alcohol, ceryl alcohol, octacosyl alcohol, melissyl alcohol, and other monohydric alcohols; ethylene glycol, 1,2-butanediol, 1,3-butanediol, 1,4-butanediol, 2,3-butanediol, and other dihydric alcohols; glycerin and other trihydric alcohols; and other polyhydric alcohols and the like.

[0029] Furthermore, examples of the abovementioned synthetic wax include stearyl laurate, stearyl myristate, stearyl stearate, octyl behenate, lauryl behenate, myristyl behenate, stearyl behenate, behenyl behenate, pentacosyl behenate, ceryl lignocerate, octacosyl lignocerate, melissyl lignocerate, stearyl cerotate, behenyl cerotate, ceryl cerotate, melissyl cerotate, ethyl montanate, ceryl montanate, glycol montanate, and the like. Also, natural waxes include montan wax, carnauba wax, beeswax, candelilla wax, rice bran wax, purified insect wax, and the like. Among these, glycol montanate, glyceride montanate, montan wax, and the like are preferred in terms of volatility and the ease with which the resin can be released from the metal rolls, and these substances are commercially available as additives used for resin, which makes them economically suitable for use.

[0030] Also, the type of oxidized polyethylene wax is not particularly limited, and the wax may be low or high viscosity. However, when taking into account the ease with which the resin can be released from the metal rolls, a partially oxidized polyethylene wax having an acid value of 1 to 40 mg KOH/g and whose molecular weight is a weight-average molecular weight of 10,000 or less is preferred. These substances are also commercially available as additives used for resin, which makes them economically suitable for use.

[0031] The polyester resin composition used for calendering according to the present invention can be manufactured simply by means of combining a fatty acid having a carbon number of 20 or higher and a metal salt of a fatty acid, and as needed, a fatty acid ester and oxidized polyethylene wax, with a pelleted or powdered polyester resin, or as a result of melt mixing these ingredients in advance in a mixer. A publicly known device may be used herein as the mixer, but rolls, a single or twin screw extruder, kneader, co-kneader, planetary mixer, Banbury mixer, or the like that is easily operated and that can achieve an even dispersion is used preferably.

[0032] Also, the polyester resin composition of the present invention can be manufactured either by means of simply mixing a fatty acid and metal salt of a fatty acid, as well as an ingredient in

which a fatty acid ester and oxidized polyethylene wax is admixed at a high concentration (usually referred to as a master batch) that is prepared in advance, into a polyester resin, or as a result of melt mixing the polyester resin with the master batch. The mixers cited above are also suitable for use in manufacturing the master batch.

[0033] The following additives may also be arbitrarily admixed in the polyester resin composition of the present invention in a range that does not harm the workability of the resin during calendering: hindered phenols, thioethers, amines, phosphoric acids, and other conventional, publicly known antioxidants; benzophenones, benzoates, benzotriazoles, cyanoacrylates, hindered amines, and other conventional, publicly known UV absorbers; anionic, cationic, or non-ionic low-molecular weight or high-molecular weight antistatic agents; epoxy compounds, isocyanate compounds, and other thickeners; dyes, pigments, and other colorants; titanium oxide, carbon black, and other UV blockers; glass fibers, carbon fibers, and other reinforcing materials; silica, clay, calcium carbonate, barium sulfate, glass beads, talc, and other fillers; flame retardants, plasticizers, antifoaming agents, antimicrobial agents, anti-mildew agents, fluorescing agents, surfactants, cross-linking agents, and the like.

[0034] Furthermore, polyolefins, polyvinyl chloride, polystyrene, polyethers, polyesters, polyamides, polyimides, and other thermoplastic resins may be admixed in the polyester resin composition of the present invention in a range that does not harm the workability of the resin during calendering.

[0035] The polyester resin composition of the present invention is suitable as a resin composition for manufacturing a sheet or film by means of a calendering machine after being melt mixed using a mixer or a single or twin-screw extruder. The molten resin can be easily released from the metal rolls and is transparent when made a sheet or film in this manner. Furthermore, longitudinal streaking due to entrainment of air in the sheet or film thus obtained does not occur, and there is no plating out to the metal rolls. Therefore, it is possible to mold a sheet or film with excellent outward appearance and high productivity by means of calendering this polyester resin composition.

[0036] A sheet or film obtained by means of calendering using the polyester resin composition pertaining to the present invention is suitable for use in transparent or colored sheeting used for decorative sheets and films, sheets or films used for films and cards, sheets or films used for various types of packaging and containers, and the like.

[Working Examples] The present invention will next be described in further detail by citing specific working examples, but the present invention is not limited by these working examples. [0038] < Working Example 1>

Kodar PETG Copolyester grade Eastar PETG 6763 (hereinafter referred to as "[A1]"), manufactured by Eastman Chemical Products, Inc., was used as the polyester resin. This polyester resin [A1] consists of pellets of a polyester resin with a number-average molecular. weight of 26,000, and comprises repeating units of a dicarboxylic acid component and a glycol component in which the dicarboxylic acid component consists of 100 mol% of terephthalic acid and no other dicarboxylic acid components, and the glycol component consists of 60 to 75 mol% of ethylene glycol, 40 to 25 mol% of 1,4-cyclohexane dimethanol, and no other glycol components. This polyester resin [A1] is also non-crystalline, with no responses observed that correspond to fusion of crystals or crystallization from a melted state when measured with the help of a differential scanning calorimeter. Montan wax (Grade Licowax S; hereinafter referred to as "[B1]"; manufactured by Clariant Japan (Inc.)) in the form of a flaked solid at room temperature was used as the fatty acid with a carbon number of 20 or higher. Also, calcium montanate (Grade Licomont Ca V102; hereinafter referred to as "[C2]"; manufactured by Clariant Japan (Inc.)) was used as the metal salt of a fatty acid. The polyester resin [A1], the fatty acid [B1] with a carbon number of 20 or above, and the metal salt [C1] of a fatty acid were admixed in a mixture ratio in which the [A1]:[B1]:[C1] weight ratio was 100:0.75:0.75; 0.5 weight part of a phenol-type antioxidant (Adekastab, grade AO-60; hereinafter referred to as "AO-60"; manufactured by Asahi Denka Co., Ltd.) was admixed therein; and a sheet was manufactured using a common calendering method. Specifically, a polyester resin composition was prepared by means of uniformly mixing the polyester resin [A1], the fatty acid [B1] with a carbon number of 20 or above, and the metal salt [C1] of a fatty acid in a Henschel mixer, and kneading the product in a Banbury mixer until the resin temperature was in a range of 160 to 175°C. The product was then rolled using a calendering machine with four reverse L-shaped rolls adjusted to a constant temperature, and the product was taken off and put through a cooling step to manufacture a sheet with a thickness of 150 µm and a width of 1,000 mm. In the sheet molding process, the release of the material (molten resin) from the calender rolls, the presence of plating out on the calender rolls, and the presence of longitudinal streaks due to entrainment of

air were evaluated, and the haze was measured as an indicator of sheet transparency by means of the reagents and methods below. Determination of each category was performed according to the evaluation criteria below. The evaluation results for the working examples are shown in Table 1, and the evaluation results for the comparative examples are shown in Table 2.

#### [0039] [Measurement of haze]

Measurement of haze was performed using a direct reading haze computer (model HGM-2D) manufactured by Suga Test Instruments (Inc.) in accordance with JIS K 7105. Five plates with dimensions of 5 cm×5 cm were cut from the central portion of a 150-μm-thick sheet obtained by means of calendering, the haze thereof was measured, and the average value thereof was taken as a representative value.

#### [0040] [Evaluation criteria]

#### (1) Evaluation of the ease of release from metal rolls

Evaluation 1 of release properties: A basic evaluation of the ease of release from metal was performed using a miniature mixer prior to molding in the calendering machine. A lab mixer (R-60) with a capacity of 60 cc manufactured by Toyo Seiki Seisakusho (Inc.) was used as the miniature mixer. At that time, the temperature of the mixer was set to 180°C, the product was kneaded for 10 minutes at 60 rpm, and the ease of release of the molten product from the mixer was evaluated. This evaluation was performed prior to calendering, and for cases in which release properties were not obtained in the evaluation, the product was considered inadequate, and further calendering thereof was not performed. For cases in which the release properties were considered good with the miniature mixer, calendering was performed, and release properties were evaluated according to the criteria below.

Evaluation 2 of release properties: Cases in which the molten sheet wound around the calender rolls could be released from the rolls set to 190°C were considered good (o), and cases in which the product adhered to the rolls were considered unsatisfactory (×).

#### (2) Evaluation of plating out to the metal rolls

Cases in which plating out to the calender rolls did not occur during two hours of continuous molding were considered good (o), and cases in which plating out occurred were considered unsatisfactory (X).

#### (3) Evaluation of longitudinal streaking due to air entrainment

Cases in which air was not entrained in the material and longitudinal streaking did not occur in the sheet obtained during rolling with the help of the calender rolls were considered good (0), and cases in which longitudinal streaking occurred in the sheet thus obtained were considered unsatisfactory (×).

#### (4) Evaluation of transparency

Cases in which the measured haze value was less than 5% were considered excellent (③), cases in which the haze value was greater than 5% and less than 10% were considered good (o), and cases in which the haze value was 10% or more were considered unsatisfactory (×). Also, the transparency of the manufactured sheet was considered good when the haze was excellent (⑤) or good (o).

#### [0041] < Working Example 2>

A polyester resin composition comprising a polyester resin [A1], a fatty acid [B1] with a carbon number of 20 or above, and a metal salt [C1] of a fatty acid was used in the same manner as described in Working Example 1. The same procedure was followed as described in Working Example 1, except that the mixture ratio of the polyester resin [A1], fatty acid [B1] with a carbon number of 20 or above, and metal salt [C1] of a fatty acid was in an [A1]:[B1]:[C1] weight ratio of 100:1.05:0.06.

#### [0042] < Working Example 3>

A polyester resin composition comprising a polyester resin [A1], a fatty acid [B1] with a carbon number of 20 or above, and a metal salt [C1] of a fatty acid was used in the same manner as described in Working Example 1. The same procedure was followed as described in Working Example 1, except that the mixture ratio of the polyester resin [A1], fatty acid [B1] with a carbon number of 20 or above, and metal salt [C1] of a fatty acid was in an [A1]:[B1]:[C1] weight ratio of 100:0.75:1.5.

#### [0043] < Working Example 4>

[A1] was used as the polyester resin, [B1] was used as the fatty acid with a carbon number of 20 or above, and zinc behenate in the form of a powdered solid at room temperature (Grade ZS-7, hereinafter referred to as [C2]; manufactured by Nippon Chemical Industries (Inc.)) was used as the metal salt of a fatty acid. The same procedure was followed as described in Working Example 1, except that the mixture ratio of the polyester resin [A1], fatty acid [B1]

with a carbon number of 20 or above, and metal salt [C2] of a fatty acid was in an [A1]:[B1]:[C2] weight ratio of 100:1.05:0.06.

#### [0044] < Working Example 5>

[A1] was used as the polyester resin, and behenic acid in the form of a powdered solid at room temperature (for use as an additive; hereinafter referred to as [B2]; manufactured by Kosei (Inc.)) was used as the fatty acid with a carbon number of 20 or above. Also, [C2] was used as the metal salt of a fatty acid. The same procedure was followed as described in Working Example 1, except that the mixture ratio of the polyester resin [A1], fatty acid [B2] with a carbon number of 20 or above, and metal salt [C2] of a fatty acid was in an [A1]:[B2]:[C2] weight ratio of 100:1.05:0.06.

#### [0045] < Working Example 6>

[A1] was used as the polyester resin, and [B1] was used as the fatty acid with a carbon number of 20 or above. Also, magnesium montanate in the form of a powdered solid at room temperature (Grade MS-8; hereinafter referred to as [C3]; manufactured by Nippon Chemical Industries (Inc.)) was used as the metal salt of a fatty acid. The same procedure was followed as described in Working Example 1, except that the mixture ratio of the polyester resin [A1], fatty acid [B1] with a carbon number of 20 or above, and metal salt [C3] of a fatty acid was in an [A1]:[B1]:[C3] weight ratio of 100:1.05:0.06.

#### [0046] < Working Example 7>

Tsunami Copolyester grade GS2 (hereinafter referred to as "[A2]"), manufactured by Eastman Chemical Products, Inc. was used as the polyester resin. This polyester resin [A2] consists of pellets of a polyester resin with a number-average molecular weight of 25,000, and comprises repeating units of a dicarboxylic acid component and a glycol component in which the dicarboxylic acid component consists of 100 mol% of terephthalic acid and no other dicarboxylic acid components, and the glycol component consists of 60 to 75 mol% of ethylene glycol, 40 to 25 mol% of 1,4-cyclohexane dimethanol, and no other glycol components. This polyester resin [A2] is also non-crystalline, with no responses observed that correspond to fusion of crystals or crystallization from a melted state when measured with the help of a differential scanning calorimeter. The same procedure was followed as described in Working Example 1, except that the mixture ratio of the polyester resin [A2], fatty acid [B1] with a carbon number of 20 or above, and metal salt [C1] of a fatty acid was in an [A2]:[B1]:[C1] weight ratio of 100:1.05:0.06.

#### [0047] < Working Example 8>

A polyester resin composition comprising a polyester resin [A1], a fatty acid [B1] with a carbon number of 20 or above, and a metal salt [C1] of a fatty acid was used in the same manner as described in Working Example 2. However, a master batch in which the fatty acid [B1] with a carbon number of 20 or above and metal salt [C1] of a fatty acid were admixed therein with high filling volumes was manufactured in advance, and a composition with the desired mixture ratio was manufactured by means of admixing a prescribed quantity of the master batch into the polyester resin [A1]. The master batch was manufactured while the mixture ratio of the polyester resin [A1], fatty acid [B1] with a carbon number of 20 or above, and metal salt [C1] of a fatty acid was kept in an [A1]:[B1]:[C1] weight ratio of 100:7.5:0.4, and a twin-screw extruder with a cylinder diameter of 35 mm was used in the manufacture thereof. The master batch thus manufactured was admixed in a prescribed quantity with respect to the polyester resin [A1] to yield an [A1]:[B1]:[C1] ratio of 100:1.05:0.06; 0.5 weight part of an antioxidant (AO-60) was admixed therein; and a sheet with a thickness of 150 µm and a width of 1,000 mm was manufactured in the same manner as described in Working Example 1.

#### [0048] < Working Example 9>

A polyester resin composition comprising a polyester resin [A2], a fatty acid [B1] with a carbon number of 20 or above, and a metal salt [C1] of a fatty acid was used in the same manner as described in Working Example 7. However, a master batch in which the fatty acid [B1] with a carbon number of 20 or above and metal salt [C1] of a fatty acid were admixed therein with high filling volumes was manufactured in advance, and a composition with the desired mixture ratio was manufactured by means of admixing a prescribed quantity of the master batch into the polyester resin [A2]. The master batch was manufactured while the mixture ratio of the polyester resin [A2], fatty acid [B1] with a carbon number of 20 or above, and metal salt [C1] of a fatty acid was kept in an [A2]:[B1]:[C1] weight ratio of 100:7.5:0.4, and a twin-screw extruder with a cylinder diameter of 35 mm was used in the manufacture thereof. The master batch thus manufactured was admixed in a prescribed quantity with respect to the polyester resin [A2] to yield an [A2]:[B1]:[C1] ratio of 100:1.05:0.06; 0.5 weight part of an antioxidant (AO-60) was admixed therein; and a sheet with a thickness of 150 µm and a width of 1,000 mm was manufactured in the same manner as described in Working Example 1.

#### [0049] < Working Example 10>

A fatty acid ester was admixed together with a polyester resin composition comprising a polyester resin [A1], a fatty acid [B1] with a carbon number of 20 or above, and a metal salt [C1] of a fatty acid in the same manner as described in Working Example 1. An ester of montanic acid and 1,4-butanediol (montan wax; Grade Licowax E; hereinafter referred to as "[D1]"; manufactured by Clariant Japan (Inc.)) was used as a fatty acid ester. The polyester resin [A1], fatty acid [B1] with a carbon number of 20 or above, metal salt [C1] of a fatty acid, and fatty acid ester [D1] were admixed in an [A1]:[B1]:[C1]:[D1] weight ratio of 100:0.75:0.04:0.04; 0.5 weight part of an antioxidant (AO-60) was admixed therein; and a sheet with a thickness of 150 µm and a width of 1,000 mm was manufactured in the same manner as described in Working Example 1.

#### [0050] < Working Example 11>

An oxidized polyethylene wax was admixed together with a polyester resin composition comprising a polyester resin [A2], a fatty acid [B1] with a carbon number of 20 or above, and a metal salt [C1] of a fatty acid in the same manner as described in Working Example 7. A product with an acid value of 15 to 19 mg KOH/g (Grade Licowax PED191; hereinafter referred to as "[D2]"; manufactured by Clariant Japan (Inc.)) was used as an oxidized polyethylene wax. The polyester resin [A2], fatty acid [B1] with a carbon number of 20 or above, metal salt [C1] of a fatty acid, and oxidized polyethylene wax [D2] were admixed in an [A2]:[B1]:[C1]:[D2] weight ratio of 100:0.75:0.04:0.04, 0.5 weight part of an antioxidant (AO-60) was admixed therein, and a sheet with a thickness of 150 µm and a width of 1,000 mm was manufactured in the same manner as described in Working Example 1.

#### [0051] < Comparative Example 1>

Units of [A1] were used as the polyester resin, and 0.5 weight part of a phenol-type antioxidant (AO-60) was added per 100 weight parts of the polyester resin [A1]. When this polyester resin composition was tested for the ease of release using a mixer, the molten resin adhered firmly to the mixer, and release was impossible. It was therefore determined that a sheet could not be manufactured using a calendering machine with a polyester resin composition comprising an antioxidant admixed into the polyester resin [A1], and calendering was not performed.

JP 2003 – 191266 A

Page 18

#### [0052] < Comparative Example 2>.

[0053] < Comparative Example 3>

Release testing was performed using a mixer in the same manner as described in Comparative Example 1, except that units of [A2] were used as the polyester resin. Because release of the molten resin from the mixer was impossible, calendering was not performed.

A polyester resin composition comprising only the polyester resin [A1] and fatty acid [B1] with a carbon number of 20 or above was used, with no metal salt of a fatty acid admixed therein. The polyester resin [A1] and fatty acid [B1] with a carbon number of 20 or above were admixed in an [A1]:[B1] weight ratio of 100:1.05; 0.5 wt% of an antioxidant (AO-60) was admixed therein; and a sheet with a thickness of 150 µm and a width of 1,000 mm was manufactured in the same manner as described in Working Example 1. Longitudinal streaks due to air entrainment occurred in this sheet. It was apparent from this comparative example that the occurrence of longitudinal streaks due to air entrainment cannot be eliminated using a polyester resin composition comprising only the polyester resin [A1] and fatty acid [B1] with a carbon number of 20 or above.

#### [0054] < Comparative Example 4>

[A1] was used as the polyester resin, [C1] was used as the metal salt of a fatty acid, and stearic acid (95% pure powdered reagent; hereinafter referred to as "[B3]"; manufactured by Kanto Chemical Co., (Inc.)) with a carbon number of less than 20 was admixed as a fatty acid into the two component, yielding a polyester resin composition. The polyester resin [A1], fatty acid [B3] with a carbon number of less than 20, and metal salt [C1] of a fatty acid were admixed in an [A1]:[B3]:[C1] weight ratio of 100:3.0:0.06; 0.5 wt% of AO-60 as an antioxidant was admixed therein; and release testing was performed thereon in the same manner as described in Comparative Example 1. As a result, release of the molten resin from the mixer was impossible, and no calendering was therefore performed.

#### [0055] < Comparative Example 5>

A polyester resin composition comprising only the polyester resin [A1] and metal salt [C1] of a fatty acid was used, with no fatty acid admixed therein. Specifically, after admixing the polyester resin [A1] and metal salt [C1] of a fatty acid in an [A1]:[C1] weight ratio of 100:1.5 and admixing 0.5 wt% of an antioxidant (AO-60) therein, release testing was performed with a

JP 2003 - 191266 A

mixer in the same manner as described in Comparative Example 1. As a result, release of the molten resin from the mixer was impossible, and no calendering was therefore performed.

#### [0056] < Comparative Example 6>

A polyester resin composition comprising the polyester resin [A1], fatty acid [B1] with a carbon number of 20 or above, and metal salt [C1] of a fatty acid the same as described in Working Example 1 was used, but the polyester resin [A1], fatty acid [B1] with a carbon number of 20 or above, and metal salt [C1] of a fatty acid were admixed in an [A1]:[B1]:[C1] weight ratio of 100:0.15:0.04. 0.5 weight part of an antioxidant (AO-60) was admixed therein, and release testing was performed with a mixer in the same manner as described in Comparative Example 1. As a result, a portion of the molten resin adhered to the mixer, and complete release of the resin was impossible. Consequently, calendering was not performed.

#### [0057] < Comparative Example 7>

A polyester resin composition comprising the polyester resin [A1], fatty acid [B1] with a carbon number of 20 or above, and metal salt [C1] of a fatty acid the same as described in Working Example 1 was used, but the polyester resin [A1], fatty acid [B1] with a carbon number of 20 or above, and metal salt [C1] of a fatty acid were admixed in an [A1]:[B1]:[C1] weight ratio of 100:3.5:0.06. After admixing 0.5 weight part of an antioxidant (AO-60) therein, a sheet with a thickness of 150 µm and a width of 1,000 mm was manufactured in the same manner as described in Working Example 1. It was apparent from this comparative example that transparency declines as the admixed quantity of the fatty acid [B1] with a carbon number of 20 or above is increased.

#### [0058] < Comparative Example 8>

The polyester resin [A1], fatty acid [B1] with a carbon number of 20 or above, and metal salt [C1] of a fatty acid were admixed in an [A1]:[B1]:[C1] weight ratio of 100:1.05:0.005, and after admixing 0.5 weight part of an antioxidant (AO-60) therein, a sheet with a thickness of 150 µm and a width of 1,000 mm was manufactured in the same manner as described in Working Example 1. It was apparent from this comparative example that the occurrence of longitudinal streaks on the surface of the sheet is not reduced when the admixed quantity of the metal salt [C1] of a fatty acid is low.

JP 2003 – 191266 A

Page 20

#### [0059] < Comparative Example 9>

The polyester resin [A1], fatty acid [B1] with a carbon number of 20 or above, and metal salt [C1] of a fatty acid were admixed in an [A1]:[B1]:[C1] weight ratio of 100:1.05:3.5, and after admixing 0.5 weight part of an antioxidant (AO-60) therein, a sheet with a thickness of 150 µm and a width of 1,000 mm was manufactured in the same manner as described in Working Example 1. It was apparent from this comparative example that the ease of release from the calender rolls is inadequate when the admixed quantity of the metal salt [C1] of a fatty acid is high, and particularly, that the release properties are inadequate when the roll temperature for smoothing the surface of the sheet is at a high temperature, and transparency declines.

#### [0060] < Comparative Example 10>

The polyester resin [A1] was used, the fatty acid ester [D1] used as described in Working Example 10 was used instead of the fatty acid, and the metal salt [C1] of a fatty acid was used. The same procedure as described in Working Example 1 was followed, except that the polyester resin [A1], fatty acid ester [D1], and metal salt [C1] of a fatty acid were admixed in an [A1]:[D1]:[C1] weight ratio of 100:1.05:0.06. It was apparent from this comparative example that the resin can be easily released from the calender rolls even when the fatty acid ester [D1] is used instead of the fatty acid [B1], but that plating out occurs.

#### [0061] < Comparative Example 11>

[A2] was used as the polyester resin, the oxidized polyethylene wax [D2] used as described in Working Example 11 was used instead of the fatty acid, and the metal salt [C1] of a fatty acid was used. The same procedure as described in Working Example 1 was followed, except that the polyester resin [A2], oxidized polyethylene wax [D2], and metal salt [C1] of a fatty acid were admixed in an [A2]:[D2]:[C1] weight ratio of 100:1.05:0.06. It was apparent from this comparative example that the resin can be easily released from the calender rolls even when the fatty acid ester [D1] is used instead of the fatty acid [B1] with a carbon number of 20 or above, but that plating out occurs.

#### [0062] < Comparative Example 12>

The polyester resin [A1] was used, polyethylene wax (grade Licowax PE520; hereinafter referred to as "[D3]"; manufactured by Clariant Japan (Inc.)) was admixed instead of the fatty acid, and the metal salt [C1] of a fatty acid was used. The polyester resin [A1], polyethylene wax [D3], and metal salt [C1] of a fatty acid were admixed in an [A1]:[D3]:[C1] weight ratio of

100:3.0:0.06, and after admixing 0.5 wt% of an antioxidant (AO-60) therein, release testing with a mixer was performed in the same manner as described in Comparative Example 1. As a result, because release of the molten resin from the mixer was impossible, calendering was not performed.

#### [0063] < Comparative Example 13>

The polyester resin [A2] was used, ethylene bis-stearyl amide (amide wax, grade Licolub FA1; hereinafter referred to as "[D4]"; manufactured by Clariant Japan (Inc.)) was admixed instead of the fatty acid, and [C1] was used as the metal salt of a fatty acid. The polyester resin [A2], ethylene bis-stearyl amide [D4], and metal salt [C1] of a fatty acid were admixed in an [A2]:[D4]:[C1] weight ratio of 100:3.0:0.06, and after admixing 0.5 wt% of an antioxidant (A0-60) therein, release testing with a mixer was performed in the same manner as described in Comparative Example 1. As a result, because release of the molten resin from the mixer was impossible, calendering was not performed.

#### [0064] < Comparative Example 14>

The polyester resin [A2] was used, a high polymer acrylic acid lubricant (Metablen, grade L-1000; hereinafter referred to as "[D5]"; manufactured by Mitsubishi Rayon (Inc.)) was admixed instead of the fatty acid, and [C1] was used as the metal salt of a fatty acid. The polyester resin [A2], high polymer acrylic acid lubricant [D5], and metal salt [C1] of a fatty acid were admixed in an [A2]:[D5]:[C1] weight ratio of 100:3.0:0.06, and after admixing 0.5 wt% of an antioxidant (AO-60) therein, release testing with a mixer was performed in the same manner as described in Comparative Example 1. As a result, because release of the molten resin from the mixer was impossible, calendering was not performed.

#### [0065] < Comparative Example 15>

[A1] was used as the polyester resin, [B1] was used as the fatty acid with a carbon number of 20 or above, and instead of the metal salt of a fatty acid, the fatty acid ester [D1] used as described in Working Example 10 was jointly admixed therein. The same procedure was followed as described in Working Example 2, except that the fatty acid ester [D1] was used instead of the metal salt [C1] of a fatty acid. It was apparent from this comparative example that when the fatty acid ester [D1] is used instead of the metal salt [C1] of a fatty acid, the occurrence of longitudinal streaks due to air entrainment is not reduced.

#### [0066] < Comparative Example 16>

[A1] was used as the polyester resin, [B1] was used as the fatty acid with a carbon number of 20 or above, and instead of the metal salt of a fatty acid, the oxidized polyethylene wax [D2] used as described in Working Example 11 was mixed with the two components. The same procedure was followed as described in Working Example 2, except that the oxidized polyethylene wax [D2] was used instead of the metal salt [C1] of a fatty acid. It was apparent from this comparative example that when the oxidized polyethylene wax [D1³] is used instead of the metal salt [C1] of a fatty acid, the occurrence of longitudinal streaks due to air entrainment is not reduced.

[**0067**] [Table 1]

				E	valuation Item	
Working Example	Composition	Mixture composition (weight	Release	Plating	Longitudinal	Transparency
		ratio)		out	streaks	
Working Example 1	[A1]/[B1]/[C1]	100/0.75/0.75	0	0	0	0
Working Example 2	[A1]/[B1]/[C1]	100/1.05/0.06	0	0	0	0
Working Example 3	[A1]/[B1]/[C1]	100/0.75/1.50	0	0	0	0
Working Example 4	[A1]/[B1]/[C2]	100/1.05/0.06	0	0	0	0
Working Example 5	[A1]/[B2]/[C2]	100/1.05/0.06	0	0	0	0
Working Example 6	[A1]/[B1]/[C3]	100/1.05/0.06	0	٥	0	©
Working Example 7	[A2]/[B1]/[C2]	100/1.05/0.06	0	0	0	0
Working Example 8	[A1]/[B1]/[C1]	100/1.05/0.06	0	0	0	©
Working Example 9	[A2]/[B1]/[C1]	100/1.05/0.06	0	0	0	0
Working Example 10	[A1]/[B1]/[C1]/[D1]	100/0.75/0.04/0.04	0	0	0	0
Working Example 11	[A1]/[B1]/[C1]/[D2]	100/0.75/0.04/0.04	0	0	0	0

<sup>&</sup>lt;sup>3</sup> Translator's note: "D1" in the original should probably read "D2," and appears to be a typographical error.

[0068] [Table 2]

			Evaluation Item			
Comparative Example	Composition	Mixture composition (weight ratio)	Release	Plating out	Longitudinal streaks	Transparency
Comparative Example 1	[A1]	100	×	-		<del> </del>
Comparative Example 2	[A2]	100	×			
Comparative Example 3	[A1]/[B1]	100/1.05	0	0	×	©
Comparative Example 4	[A1]/[B3]/[C1]	100/3.0/0.06	×			<del> </del>
Comparative Example 5	[A1]/[C1]	100/1.5	×			1
Comparative Example 6	[A1]/[B1]/[C1]	100/0.15/0.04	×			
Comparative Example 7	[A1]/[B1]/[C1]	100/3.5/0.06	0	0	. 0	×
Comparative Example 8	[A1]/[B1]/[C1]	100/1.05/0.005	0	0	×	0
Comparative Example 9	[A1]/[B1]/[C1]	100/1.05/3.5	×	0	0	×
Comparative Example 10	[A1]/[D1]/[C1]	100/1.05/0.06	0	×	0	0
Comparative Example 11	[A2]/[D2]/[C1]	100/1.05/0.06	0	×	0	0
Comparative Example 12	[A1]/[D3]/[C1]	100/3.0/0.06	×			
Comparative Example 13	[A2]/[D4]/[C1]	100/3.0/0.06	×			
Comparative Example 14	[A2]/[D5]/[C1]	100/3.0/0.06	×			
Comparative Example 15	[A1]/[B1]/[D1]	100/1.05/0.06	0	0	×	0
Comparative Example 16	[A1]/[B1]/[D2]	100/1.05/0.06	0	0	×	- 0

<sup>\*</sup> For empty spaces in the table, because release properties were determined by means of release testing with a mixer, and calendering was not performed, there are no corresponding results for those evaluation items.

#### [0069]

[Effect of the Invention] As is apparent from the evaluation shown in Tables 1 and 2, by means of the polyester resin composition of the present invention, a polyester resin (non-crystalline polyester resin) with reduced crystallinity is provided, wherein virtually no adhesion to the metal rolls (calender rolls) or plating out during calendering occurs, the transparency of the sheet/film thus obtained is good, longitudinal streaking due to entrainment of air in the sheet or film does not occur, and the composition is suitable for calendering of sheets and films with a high rate of production.

## Continued from front page:

(51) Int. Cl. <sup>7</sup>	Class. Symbols		FI			Subject Codes (Reference)	
B 29 L 7:00				В	29 L 3	7:00	
F terms (reference):	4F071	AA45	AA46	AA47	AC09	AEII	
		AH04	AH05	BB04	BC01		
	4F204	AA24	AB07	AB19	FA07	FB02	
		FF01	FF06				
	4J002	CF031	CF041	CF051	CF061		
		CF071	CF081	CF091	EF056		
		EG027	EG037	EG047	FD176		
		FD177	GB00	GG00			